Homogeneous nucleation: Comparison between two theories

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Abstract

The classical nucleation theory of Becker, Döring and Zeldovich is compared with the Langer coarse-grained field approach to the nucleation phenomenon. Both formalisms have been applied to the condensation from a supersaturated vapor. It is shown that the nucleation rate derived in the classical theory can be expressed in a form equivalent to that of the field nucleation theory. This equivalence serves as an explanation of the puzzling fact that the numerical predictions of both theories for condensation of Xe and CO_2 are almost identical though the standard analytical expressions for the nucleation rates are different. The results obtained can help to link the theories of nucleation and their approximations.

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First order phase transitions, which include the thoroughly studied condensation of a vapor, boiling of a liquid or crystallization of a melt, as well as the hypothetical transition from a hadronic phase to the quark−gluon plasma, play an important role in modern science and technology. The theory of first order phase transitions is based on the homogeneous nucleation theory which aims to determine the rate of the decay of a metastable state, i.e. the rate of formation of nucleating clusters within the initially homogeneous nonequilibrium state. This is the first important assumption of the nucleation theory which implies that the conversion of a metastable state into a thermodynamically stable phase should proceed via the formation of clusters of the new phase. As a typical example one may consider the condensation of small drops of liquid from the vapor rapidly quenched into the metastable region.

Clusters of various sizes are produced spontaneously due to the fluctuations in a homogeneous medium. Clusters with sizes larger than a certain size, called critical, will grow into a new phase, and thus the phase transition process develops. A description of this process requires a kinetic analysis of the evolution of the clusters. The phenomenological nucleation theory, often referred to as the classical or Becker−Döring−Zeldovich (BDZ) theory, was formulated in the pioneering works by Becker and Döring [1], and Zeldovich [2]. It is explained in detail in many textbooks and reviews, see e.g. Refs. [3–9]. Despite the phenomenological origin, the classical theory appears to predict nucleation phenomena very well.

A field theoretical approach to the problem of nucleation has been proposed by Langer [10–12] who extended the generalization of the BDZ theory for a system of arbitrary many degrees of freedom [13] to the field theories. This formalism has been applied to several nucleation phenomena, including the liquid−vapor phase transition [12]. In spite of all the statistical and hydrodynamic corrections, it appears that the nucleation rate calculated within the coarse-grained field theory differs only slightly from that of the BDZ theory, while the expressions for the rates are different. This surprisingly good agreement between the two theories leads to the assumption that, although the coarse-grained field theory is a more accurate and advanced treatment of the nucleation phenomena than the classical theory, the latter compensates somehow correctly for all degrees of freedom [7,8,12].

In the present paper we would like to show that the nucleation rate derived in the BDZ theory may be obtained under certain assumptions in the form which is identical to that of the Langer theory. We will start from a brief review of the classical nucleation theory. Then, after discussion of the formalism of the coarse-grained field theory, the results of both models will be applied to condensation from a supersaturated vapor. The comparison will be done in terms of the Langer theory.

The metastable state typically is assumed to be a mixture of molecules and clusters of the new phase containing n molecules. Let $N_n(t)$ be the average number of clusters of size n at time t. Effects of the direct interactions between the clusters are disregarded. Therefore, the clusters may change their size only due to an evaporation$-$condensation mechanism, in which a cluster of size n grows or shrinks by condensation or loss of a single molecule. This is the basic assumption of the classical nucleation theory. The rate of change of the number of clusters of a given size is determined by the master equation

$$\frac{\partial N_n(t)}{\partial t} = J_n - J_{n-1} \ , \ n \geq 2 \ , \tag{1}$$

where

$$J_n = g_{n-1}^+ \, N_{n-1}(t) - g_n^- \, N_n(t) \tag{2}$$

is a current in a size space, i.e. it is the rate at which clusters of size $n-1$ grow to clusters of size n. Here g_{n-1}^+ is the rate of the molecule condensation to a cluster of size n, and g_n^- is the rate of evaporation. According to the theory of thermodynamic fluctuations the equilibrium distribution of clusters obeys the Boltzmann (or Gibbs) distribution

$$N_n^{eq} = N_0^{eq} \exp\left(-\frac{\Delta G_n}{k_B T}\right) \ , \tag{3}$$

where ΔG_n is the minimum work needed to form a cluster of size n, k_B is Boltzmann's constant, T is the temperature, and N_0^{eq} is a pre-exponential factor. A maximum in ΔG_n is reached at $\partial \Delta G_n / \partial n = 0$ corresponding to a cluster of critical size, n_c. Clusters smaller than the critical cluster tend to shrink, clusters of the critical size are in a metastable equilibrium, and clusters larger than the critical cluster tend to increase their volume.

The equilibrium cluster distribution function N_i^{eq} is a time independent solution of the master equation (1). The current J_n is zero for this distribution, therefore, one may write the equation of detailed balance

$$g_{n-1}^+ \exp\left(-\frac{\Delta G_{n-1}}{k_B T}\right) = g_n^- \exp\left(-\frac{\Delta G_n}{k_B T}\right) \ . \tag{4}$$

As shown by Zeldovich [2], under the treatment of n as a continuous variable the master equation (1) may be approximated by the Fokker$-$Planck kinetic equation

$$\frac{\partial N_n(t)}{\partial t} = -\frac{\partial J_n}{\partial n} = \frac{\partial}{\partial n}\left[\frac{g_n}{k_B T}\frac{\partial \Delta G_n}{\partial n}N_n(t) + g_n\frac{\partial N_n(t)}{\partial n}\right] \ , \tag{5}$$

or, in terms of the ratio $N_n(t)/N_n^{eq}$,

$$\frac{\partial N_n(t)}{\partial t} = \frac{\partial}{\partial n}\left[g_n N_n^{eq} \frac{\partial}{\partial n}\left(\frac{N_n(t)}{N_n^{eq}}\right)\right] . \tag{6}$$

The classical theory is based on a nonequilibrium, steady-state solution of Eq. (5), corresponding to a continuous phase transition. Then $J_n = s = constant$, which is called the nucleation rate and measures the number of clusters passing through the critical range per unit time per unit volume.

The boundary conditions for small and large n are chosen in the BDZ theory as follows,

$$\lim_{n\to 0}\frac{N_n(t)}{N_n^{eq}} = 1 \quad , \quad \lim_{n\to\infty}\frac{N_n(t)}{N_n^{eq}} = 0 . \tag{7}$$

A number of small clusters is assumed to be close to its equilibrium value due to the rapid increase of the fluctuation probability with decrease of cluster size. It is assumed also that clusters larger than that of critical size are removed from the system. The steady-state solution of Eqs. (6) and (7) reads

$$\frac{N_n(t)}{N_n^{eq}} = s \int_n^\infty \frac{dn'}{g'_n N_{n'}^{eq}} , \tag{8}$$

where

$$s^{-1} = \int_0^\infty \frac{dn'}{g'_n N_{n'}^{eq}} . \tag{9}$$

Since the function $N_n^{eq} = N_0^{eq}\exp(-\Delta G_n/k_B T)$ has a very sharp maximum at n_c, the integral in (9) can be evaluated by the saddle-point method, i.e. by expanding of ΔG_n around n_c and extending the integration with respect to $n - n_c$ from $-\infty$ to ∞. With these approximations, the result for the steady-state nucleation rate may be written in the general form

$$s = g_{n_c} N_{n_c}^{eq} Z , \tag{10}$$

containing the Zeldovich factor

$$Z = \left(\frac{\gamma}{2\pi k_B T}\right)^{1/2} \quad , \quad \gamma = -\left(\frac{\partial^2 \Delta G_n}{\partial n^2}\right)_{n=n_c} . \tag{11}$$

4

Here the number of clusters that have passed through the critical range is expressed via the equilibrium number of clusters of critical size. This is the principal result of the classical theory.

The accretion rate of the molecule condensation to the surface of the critical cluster g_{n_c} is derived from the kinetic theory of gases to be

$$g_{n_c} = \frac{p\, S_c}{\sqrt{2\pi k_B T m_1}} \; , \tag{12}$$

containing the gas pressure p, the surface of the critical droplet S_c, and the average mass of a single molecule m_1. On the other hand g_{n_c} may be obtained from the solution of rather general equations describing the diffusion growth of dense clusters. We will come to this point later during the discussion of vapor condensation.

An extension of the classical theory to a system of arbitrary many degrees of freedom was worked out by Landauer and Swanson [13]. Their method was applied by Langer to the field nucleation theory [11]. The BDZ and the Langer theories postulate that unstable fluctuations of thermal origin, e.g. clusters of critical size, lead to the decay of the metastable state. However, in the field nucleation theory the critical cluster needs not necessarily be a physical object [14], but rather characterizes a certain saddle-point configuration. In contrast to the classical theory, only the critical cluster and its shape deformations are required for the field theory of nucleation. The advantages and shortcomings of both models are discussed in detail in [7].

The Langer theory starts with the consideration of a classical system with $\mathcal{N}$ degrees of freedom (coordinates and momenta) η_i, $i = 1, \ldots, \mathcal{N}$. The probability distribution functional $\rho(\{\eta_i\}, t)$ is assumed to obey a continuity equation in $\{\eta\}$-space, namely

$$\frac{\partial \rho(\{\eta_i\}, t)}{\partial t} = -\sum_{i=1}^{\mathcal{N}} \frac{\partial J_i}{\partial \eta_i} \; , \tag{13}$$

which is analogous to the multi-dimensional Fokker$-$Planck equation. The probability current J_i is given by

$$J_i = -\sum_{i=1}^{\mathcal{N}} M_{ij} \left(\frac{\partial F}{\partial \eta_i} \rho + k_B T \frac{\partial \rho}{\partial \eta_i} \right) \; , \tag{14}$$

containing a generalized mobility matrix M_{ij}, and a coarse-grained free energy functional $F\{\eta\}$. For the case of $J_i = 0$ the equilibrium solution of Eq. (13)

reads

$$\rho_{eq}\{\eta_i\} \propto \exp\left(-\frac{F\{\eta_i\}}{k_B T}\right) \ . \tag{15}$$

The basic idea of the field nucleation theory is that the states of metastable and stable equilibrium lie in the vicinity of the local minima of $F\{\eta_i\}$ in the $\{\eta\}$-space, which maximize $\rho_{eq}\{\eta_i\}$. The phase transition occurs when the configuration $\{\eta_i\}$ moves from the vicinity of a metastable minimum to the vicinity of a stable minimum. To develop into the stable state the configuration must overcome the potential barrier. It is most likely for the trajectory of the system to pass across a small area around the intermediate saddle point of the functional F, say $\{\bar{\eta}\}$. This saddle-point configuration $\{\bar{\eta}\}$ corresponds to the critical cluster of a condensing phase in the BDZ theory, although it may not be a physical object. The rate of the decay of the metastable state is determined by the steady-state solution of Eqs. (13) and (14), corresponding to a finite probability current across the saddle point from the metastable to the stable minimum of F.

To obtain this solution Langer [11] introduced a set of new variables ξ_k, $k = 1,\ldots,\mathcal{N}$, whose fluctuations are slow compared to the characteristic time of the evaporation−condensation process. These variables should contain the order parameter. They are related to the η's by virtue of the orthogonal transformation $\hat{D}$:

$$\xi_j = \sum_i D_{ij}\left(\eta_i - \bar{\eta}_i\right) \ , \tag{16}$$

where, near the saddle point $\{\xi\} = \{0\}$,

$$F\{\eta\} = \bar{F} + \frac{1}{2}\sum_{j=1}^{\mathcal{N}} \lambda_j \xi_j^2 + \ldots \tag{17}$$

Here $\bar{F} = F\{\bar{\eta}\}$, and λ_j are the eigenvalues of the matrix $M_{ij} = (\partial^2 F/\partial\eta_i \partial i\eta_j)_{\eta=\bar{\eta}}$. By the definition of the saddle point at least one of the eigenvalues λ_j must be negative. Following Langer we denote this eigenvalue as λ_1. Then in terms terms of the ratio $\sigma(\{\xi\}) = \rho(\{\xi\})/\rho_{eq}(\{\xi\})$ the requirement of a steady state current over the potential barrier has the form

$$\sum_i \frac{\partial J_i}{\partial \xi_i} = -\sum_{ij} M_{ij} \frac{\partial}{\partial \xi_i}\left(\rho_{eq}(\{\xi\})\frac{\partial \sigma(\{\xi\})}{\partial \xi_j}\right) = 0 \ , \tag{18}$$

which is a straightforward generalization of Eq. (6). The stationary probability distribution functional $\rho(\{\xi\})$ obeys the boundary conditions that, near the

metastable state, $\rho(\{\xi\})$ is very close to the equilibrium distribution $\rho_{eq}\{\eta_i\}$, and beyond the saddle point $\rho(\{\xi\})$ should vanish rapidly. The solution of Eq. (18) which satisfies these boundary conditions becomes

$$\sigma(\{\xi\}) = \frac{1}{\sqrt{2\pi k_B T}} \int\limits_{u}^{\infty} \exp\left(-\frac{a^2}{2 k_B T}\right) da \ , \tag{19}$$

where u is a linear combination of the ξ's

$$u = \sum_i U_i \xi_i \ . \tag{20}$$

The coefficients U_i are solutions of the eigenvalue equation

$$\lambda_n \sum_i M_{ni} U_i = -\kappa U_i \ , \tag{21}$$

which describe the growth rate κ of the single unstable mode at the saddle point. To obtain the desired nucleation rate one has to integrate the flux J_i over any plane containing the saddle point and not parallel to J_i. After the cumbersome but straightforward calculations the steady state solution of Eq. (18) may be written in the form [11]

$$I = I_0 \exp\left(-\frac{\Delta F_c}{k_B T}\right) \ , \tag{22}$$

where

$$I_0 = |\kappa| \, \mathcal{V} \left(\frac{k_B T}{2\pi |\lambda_1|}\right)^{1/2} = \frac{|\kappa|}{2\pi} \Omega_0 \ . \tag{23}$$

Here $\Delta F_c = \bar{F}(\bar{\eta}) - \bar{F}(\eta_0)$ is the excess free energy required to form a cluster of the critical size. The dynamical prefactor κ describes the cluster growth at the saddle point, the statistical prefactor Ω_0 is often called the generalization of the Zeldovich factor, and $\mathcal{V}$ denotes the volume of the saddle point subspace. Although Eq. (22) has a structure similar to Eq. (10), it is asserted that the prefactors in both equations should be different [12]. However, up to now there is no crucial experiment which can distinguish the predictions of Langer theory from those of the BDZ theory.

To compare the models let us consider the condensation of a supersaturated vapor. The minimum work needed to form a spherical droplet of radius R in

the thin wall approximation is simply

$$\Delta F(R) = -\frac{4\pi}{3}R^3 \Delta p + 4\pi R^2 \sigma \; , \tag{24}$$

where Δp is the difference between the pressures inside and outside the droplet and σ is the surface tension. The critical radius is given by the Laplace formula $R_c = 2\sigma/\Delta p$, and near this value $\Delta F(R)$ can be represented by harmonic approximation

$$\Delta F(R) = \frac{4}{3}\pi\sigma R_c^2 - 4\pi\sigma(R - R_c)^2 \; . \tag{25}$$

It is convenient to rewrite the excess of the free energy in terms of the reduced radius $r = R/R_c$ and the new similarity number $\lambda_Z = R_c\sqrt{\dfrac{4\pi\sigma}{k_B T}}$ introduced by the authors [15] recently,

$$\Delta F(R) = \frac{k_B T}{3}\lambda_Z^2 - k_B T(r - 1)^2 \lambda_Z^2 \tag{26}$$

From the definition (17) it follows that $\xi = r - 1$, the free energy of the critically large droplet is $\bar{F} = k_B T \lambda_Z^2/3$ and the only negative eigenvalue λ_1 is $\lambda_1 = 2k_B T \lambda_Z^2$. Therefore, the nucleation rate predicted by the Langer theory becomes

$$I_0^L = \frac{|\kappa|}{2\sqrt{\pi}}\frac{\mathcal{V}}{\lambda_Z} \; . \tag{27}$$

The statistical prefactor Ω_0 is obviously determined by the formula

$$\Omega_0 = \mathcal{V}\left(\frac{2\pi k_B T}{|\lambda_1|}\right) = \mathcal{V}\frac{\sqrt{\pi}}{\lambda_Z} \; . \tag{28}$$

Substituting Eq. (26) in Eq. (11) we get for the Zeldovich factor

$$\gamma = -\left(\frac{\partial^2 \Delta F}{\partial R^2}\right)_{R_c} = 2k_B T\left(\frac{\lambda_Z}{R_c}\right)^2 \tag{29}$$

and

$$Z = \left(\frac{\gamma}{2\pi k_B T}\right)^{1/2} = \frac{\lambda_Z}{\sqrt{\pi}R_c} \; . \tag{30}$$

8

The factor Z has the physical meaning of a measure of the width Δ of the Gaussian distribution by which the actual distribution $\rho^{-1}(R)$ is approximated, namely $Z = \Delta^{-1}$. Therefore, the statistical prefactor Ω_0, which measures the volume of the saddle point in the phase space, can not be considered as a simple generalization of the Zeldovich factor. Instead, it is related to the Z factor by

$$\Omega_0 = \frac{\mathcal{V}}{R_c} Z^{-1} . \tag{31}$$

The nucleation rate derived in the BDZ theory is

$$I_0^{BDZ} = g(R_c) \frac{\lambda_Z}{\sqrt{\pi} R_c^2} \mathcal{V} , \tag{32}$$

where one has to evaluate the rate $g(R_c)$. From the significance of the current J, the drift coefficient $\dfrac{g}{k_B T} \dfrac{\partial \Delta F}{\partial R}$ acts as a velocity in size space [6], dR/dt, which can be determined independently by the solution of the macroscopic equations for the growth of a droplet. We will consider the diffusion model of droplet growth which assumes that the droplet of a new phase grows due to the diffusion flux of matter through its interface. Then, the problem of the liquid$-$gas phase transition is similar to the case of precipitation of a substance from a supersaturated solution or separation in binary fluids. The only difference is that one has to choose the order parameter as the local entropy density in a liquid$-$gas system, and as the local concentration in a weakly supersaturated solution [17]. For the latter case the spherically symmetric concentration distribution $c(r)$ around a droplet of radius R is given by the steady state solution of the diffusion equation

$$D\nabla^2 c(r) = \frac{\partial c(r)}{\partial t} = 0 , \tag{33}$$

where D is the solute diffusion coefficient. We obtain

$$c(r) = c - (c - c_{0R})\frac{R}{r} , \tag{34}$$

where

$$c_{0R} = c_0 + \frac{R}{r}(c - c_0) \tag{35}$$

is the equilibrium concentration of the solution at the surface of a droplet of radius R, c_0 is the equilibrium concentration of the solution above the planar

surface, and c is the given value of the concentration of the supersaturated solution. Substituting Eq. (35) in Eq. (34) and identifying δc as the supersaturation $c - c_0$ in the system, we get

$$c(r) = c - \delta c \frac{R_c}{R} \left(1 - \frac{R_c}{R} \right) \ . \tag{36}$$

Since the diffusive flux into the droplet $i(r) = D\dfrac{\partial c}{\partial r}$ at a droplet surface is equal to the growth rate of the droplet, one may write

$$\frac{dR}{dt} = \frac{D\,\delta c}{R} \left(1 - \frac{R_c}{R} \right) \ . \tag{37}$$

Linearizing this equation around the critical radius and taking into account that, by definition of the dynamical prefactor, $R - R_c = \exp(-\kappa t)$, we have finally

$$\frac{dR}{dt} = |\kappa| \left(\frac{R_c}{R} \right)^2 (R - R_c) \ , \tag{38}$$

where $|\kappa| = \dfrac{D\,\delta c}{R^2}$. This radial dependence of the droplet growth rate is valid for the liquid$-$gas phase transition also. Similar result was obtained in Ref. [16] by virtue of the hydrodynamic Kotchine equations describing the enlargement of a droplet. Thus, for the critically large droplet we have

$$g(R_c) = k_B T \left(\frac{dR}{dt} \right)_{R_c} \left[\frac{\partial \Delta F}{\partial R} \right]_{R_c}^{-1} = \frac{|\kappa|}{2\lambda_Z^2} R_c^2 \ . \tag{39}$$

Substitution of Eq. (39) in Eq. (32) yields

$$I_0^{BDZ} = \frac{|\kappa|}{2\sqrt{\pi}} \frac{\mathcal{V}}{\lambda_Z} \ , \tag{40}$$

leading again to the result (27).

For the sake of simplicity we considered the process where the one order parameter was sufficient for the description of nucleation. Obviously, our analysis may be extended to the system of arbitrary many degrees of freedom. We proposed a direct method allowing one to link the classical theory to the modern field theory of nucleation. It is very important for the comparison that the underlying dynamics of the process, that governs the cluster growth, should be the same. Trying to compare both theories Langer and Turski found [12],

10

that the numerical predictions of the critical supersaturation in Xe and CO_2 given by the field nucleation theory are very close to those of the classical theory. But the rate of molecule accretion on the surface of the critical cluster, $g(R_c)$, was calculated from simple geometric considerations of the kinetic theory of gases. Therefore, the analytical expressions of the nucleation rates were different, and it was difficult to find conformity between them.

Recently [18] the Langer theory of nucleation has been applied to calculate quark$-$gluon plasma formation in heavy ion collisions at the energy of the Brookhaven accelerator. Then the nucleation rate derived in the field theory has been compared with that of the classical theory. Again it turns out that both prefactors are about the same order of magnitude.

The classical nucleation theory is attractive not only because it predicts nucleation rates in terms of macroscopic quantities. It can be easily generalized to time-dependent nucleation phenomena. Also, the classical theory can be extended to the temperature and curvature dependences of the interfacial energy, to the deviations of the shape of the clusters from the ideal spherical shape, etc. In spite of these advantages, and although the classical theory appears to be consistent with the experimental data, it is implied explicitly and implicitly that one must consider the classical theory only as a crude approximation. This problem becomes clearer now. Using the macroscopic diffusion model for the description of a spherically growing droplet of liquid we have shown that the classical nucleation rate can be derived in a form identical to that of the field nucleation theory. The results obtained in this paper support the validity of the phenomenological classical theory on the basis of the more accurate Langer first-principles theory of nucleation. A further comparison between these theories may be useful for the development of a more rigorous nucleation theory.

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